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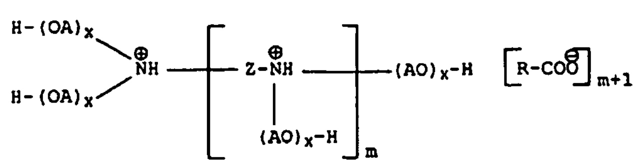
(54) Title: GASOLINEN COMPOSITIONS



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(57) Abstract: The invention provides a gasoline composition comprising a major amount of a gasoline suitable for use in a spark ignition engine; 5 to 1000 ppmw, based on total composition, of a fatty acid salt of an alkoxylated oligoamine of general formula (I) wherein each moiety A independently represents an alkylene group of 2 to 8 carbon atoms, each moiety R independently represents a C_{7-23} alkyl or singly or multiply-unsaturated C_7 to C_{23} alkenyl group, optionally substituted by one or more -OH groups, each moiety Z independently represents a C_{1-8} alkylene group, a C_{3-8} cycloalkylene group, or a C_{6-12} arylene or arylalkylene group, m represents 0 or an integer in the range 1 to 5, and the total of all variables x has a value of 50% to 300% of (m+3); and 600 to 2000 ppmw, based on total composition, of a co-additive selected from the group consisting of (a) a nitrogen-containing detergent containing a hydrocarbyl group having a number average molecular weight (Mn) in the range 750 to 6000, (b) a polyalphaolefin having a viscosity at 100 °C in the range $2x10^{-6}$ to $2x10^{-5}$ m^2/s (2 to 20 centistokes), being an oligomer containing 18 to 80 carbon atoms derived from at least one alphaolefinic monomer containing from 8 to 16 carbon atoms; (c) a polyoxyalkylene compound selected from glycols, mono- and diethers thereof, having number average molecular weight (Mn) in the range 400 to 3000; and mixtures of any two or all three of (a), (b) and (c).

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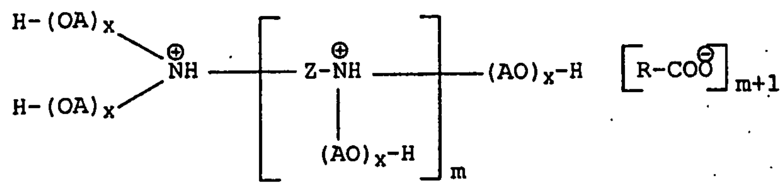
GASOLINE COMPOSITIONS

This invention relates to gasoline compositions, more particularly to such compositions containing a fatty acid salt of an alkoxylated oligoamine, and their preparation.

US Patent 4,131,583 discloses certain fatty acid salts of alkoxylated oligoamines and their use as corrosion inhibitors in aqueous coating agents for metal surfaces.

DE-A-19955651 (BASF) discloses the use of fatty acid salts of alkoxylated oligoamines of general formula I



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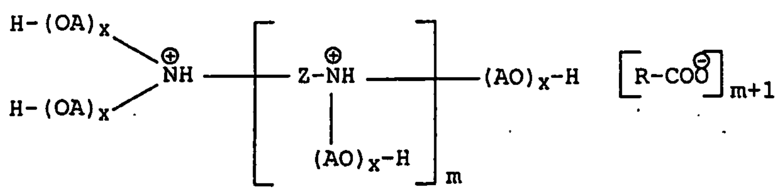
wherein each moiety A independently represents an alkylene group of 2 to 8 carbon atoms, each moiety R independently represents a C_{7-23} alkyl or singly or multiply-unsaturated C_7 to C_{23} alkenyl group, optionally substituted by one or more -OH groups, each moiety Z independently represents a C_{1-8} alkylene group, a C_{3-8} cycloalkylene group or a C_{6-12} arylene or arylalkylene group, m represents 0 or an integer in the range 1 to 5, and the total of all variables x has a value of 50% to 300% of (m+3), as lubricity improvers for mineral oil products, including fuels and lubricating oils. Use in gasoline and in diesel fuels are specifically disclosed. Use is particularly described together with a polyisobutylene amine detergent and a polyether carrier oil therefor, and Example 3 in DE-A-19955651 specifically

describes the advantageous lubricity properties of a composition based on a 50% distillation residue of a standard Eurosuper gasoline into which was incorporated 50 mg/kg (ppmw) of a specific salt of formula I and 500 mg/kg (ppmw) of an additive package containing a polyisobutylene amine detergent, a synthetic carrier oil and a conventional corrosion inhibitor. These lubricity properties are demonstrated in a high frequency reciprocating rig (HFFR) test.

It has now surprisingly been found that if a fatty acid salt of formula I is incorporated in a gasoline composition together with higher concentrations of certain co-additives than are disclosed in DE-A-19955651, significantly enhanced acceleration performance can be achieved from a spark-ignition engine fuelled by the gasoline composition.

According to the present invention therefore, there is provided a gasoline composition comprising a major amount of a gasoline suitable for use in a spark ignition engine; 5 to 1000 ppmw, based on total composition, of a fatty acid salt of an alkoxylated oligoamine of general formula I



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wherein each moiety A independently represents an alkylene group of 2 to 8 carbon atoms, each moiety R independently represents a C_{7-23} alkyl or singly or multiply-unsaturated C_7 to C_{23} alkenyl group, optionally substituted by one or more -OH groups, each moiety Z independently represents a C_{1-8} alkylene group, a C_{3-8} cycloalkylene group, or a C_{6-12} arylene or arylalkylene

group, m represents 0 or an integer in the range 1 to 5, and the total of all variables x has a value of 50% to 300% of (m+3); and 600 to 2000 ppmw, based on total composition, of a co-additive selected from the group consisting of (a) a nitrogen-containing detergent containing a hydrocarbyl group having a number average molecular weight (Mn) in the range 750 to 6000, (b) a polyalphaolefin having a viscosity at 100°C in the range $2x10^{-6}$ to $2x10^{-5}$ m^2/s (2 to 20 centistokes), being an oligomer containing 18 to 80 carbon atoms derived from at least one alphaolefinic monomer containing from 8 to 16 carbon atoms; (c) a polyoxyalkylene compound selected from glycols, mono- and diethers thereof, having number average molecular weight (Mn) in the range 400 to 3000; and mixtures of any two or all three of (a), (b) and (c).

In formula I, although A may represent any C_{2-8} alkylene group, it is preferably derived from an alkylene oxide, and most preferably represents a 1,2-ethylene, 1,2-propylene or 1,2-butylene group.

R is preferably a linear group, and preferably represents a singly or multiply-unsaturated C_{15-19} alkenyl group, optionally substituted by one or more -OH groups. The fatty acid from which the salt is derived may very conveniently be oleic acid, linoleic acid, linolenic acid, or a mixture of fatty acids containing any two or all three of such acids, e.g. tall oil fatty acid or rapeseed oil fatty acid.

Z preferably represents a polymethylene group of formula $-(CH_2)_n-$, where n is 2 to 8, more preferably 2 to 6, either as single entities (e.g. an alpha, omega-alkylene group) or a mixture of alpha, omega-alkylene groups.

The variable m preferably represents 1 or 2, more preferably 1.

The total of all variables x preferably has a value of 75% to 125% of (m+3). The value of each x is most preferably 1.

Accordingly, in formula I, it is preferred that each moiety A independently represents a 1,2-ethylene, 1,2-propylene or 1,2-butylene group, (most preferably a 1,2-propylene group) each moiety R independently represents a singly or multiply-unsaturated C_{15-19} alkenyl group, optionally substituted by one or more -OH groups (R-COOH most preferably being oleic acid, linoleic acid or a mixture thereof), each moiety is a polymethylene group of formula $-(CH_2)_n-$, where n is 2 to 6, m is 1 or 2, and the total of all variables x has a value of 75% to 125% of (m+3) (most preferably each x being 1).

The fatty acid salts of formula I may be synthesised as described in DE-A-19955651.

The nitrogen-containing detergent (a) containing a hydrocarbyl group having a number average molecular weight (Mn) in the range 750 to 6000 may be an amine, e.g. a polyisobutylene mono-amine or polyamine, such as a polyisobutylene ethylene diamine, or N-polyisobutenyl-N',N'-dimethyl-1,3-diaminopropane, or amides, e.g. a polyisobutenyl succinimide, and are variously descsribed, for example, in US Paent 5,855,629 and WO 0132812.

A particularly preferred nitrogen-containing detergent is hydrocarbyl amine of formula R^1-NH_2 and R^1 represents a group R^2 or a group R^2-CH_2- and R^2 represents a hydrocarbyl group having a number average molecular weight in the range 750 to 6000, preferably in the range 900 to 3000, more preferably 950 to 2000, and most preferably in the range 950 to 1350, e.g. a polybutenyl or polyisobutenyl group having a number average molecular weight in the range 950 to 1050.

The nitrogen-containing detergents (a) are known materials and may be prepared by known methods or by methods analogous to known methods. For example, US Patent 4,832,702 describes the preparation of polybutenyl and polyisobutenyl amines from an appropriate polybutene or polyisobutene by hydroformylation and subsequent amination of the resulting oxo product under hydrogenating conditions.

Suitable hydrocarbyl amines are obtainable from BASF A.G., under the trade mark "Kerocom".

The polyalphaolefins (b) are primarily trimers, tetramers and pentamers, and synthesis of such materials is outlined in Campen <u>et</u> <u>al</u>. "Growing use of synlubes", Hydrocarbon Processing, February 1982, Pages 75 to 82. The polyalphaolefin may be unhydrotreated, but it is preferably a hydrogenated oligomer. The polyalphaolefin (b) is preferably derived from an alphaolefinic monomer containing from 8 to 12 carbon atoms. Furthermore, it preferably has viscosity at 100°C in the range $6x10^{-6}$ to $1x10^{-5}$ m^2/s (6 to 10 centistokes). Polyalphaolefins derived from decene-1 have been found to be very effective. Polyalphaolefin having a viscosity at 100°C of $8x10^{-6}$ m^2/s (8 centistokes) has been found to be very effective.

The polyoxyalkylene compound (c) preferably has the formula II

$$R^3 - O - \left(R^5 \right)_p - O - R^4 \qquad \text{(II)}$$

wherein R^3 and R^4 independently represent hydrogen atoms or hydrocarbyl, preferably C_{1-40} hydrocarbyl, e.g. alkyl, cycloalkyl, phenyl or alkyl-phenyl groups, each R^5 independently represents an alkylene, preferably C_{2-8}

alkylene, group, and p is such that Mn of the polyoxyalkylene compound is in the range 400 to 3000, preferably 700 to 2000, more preferably 1000 to 2000.

Preferably R^3 represents a C_{8-20} alkyl group and R^4 represents a hydrogen atom. R^3 preferably represents a C_{8-18} alkyl group, more preferably a C_{8-15} alkyl group. R^3 may conveniently be a mixture of C_{8-15} alkyl groups.

In the formula II the groups R^5 are preferably 1,2 alkylene groups. Preferably each group R^5 independently represents a C_{2-4} alkylene group, e.g. an ethylene, 1,2-propylene or 1,2-butylene group. Very effective results have been obtained when each group R^5 represents a 1,2-propylene group.

Number average molecular weights, e.g. of hydrocarbons such as polyalkenes, may be determined by several techniques which give closely similar results. Conveniently Mn may be determined by vapour phase osmometry (VPO) (ASTM D 3592) or by modern gel permeation chromatography (GPC), e.g. as described for example in W.W. Yau, J.J. Kirkland and D.D. Bly, "Modern Size Exclusion Liquid Chromatography", John Wiley and Sons, New York, 1979. Where the formula of a compound is known, the number average molecular weight can be calculated as its formula weight.

Typical of gasolines suitable for use in spark ignition engines are mixtures of hydrocarbons having boiling points in the range from 25°C to 232°C and comprising mixtures of saturated hydrocarbons, olefinic hydrocarbons and aromatic hydrocarbons. Preferred are gasoline blends having a saturated hydrocarbon content ranging from 40 to 80 per cent volume, an olefinic hydrocarbon content ranging from 0 to 30 per cent volume and an aromatic hydrocarbon content ranging from 10 to 60

per cent volume. The gasoline can be derived from straight run gasoline, polymer gasoline, natural gasoline, dimer- or trimerised olefins, synthetically produced aromatic hydrocarbon mixtures from thermally or catalytically reformed hydrocarbons, or from catalytically cracked or thermally cracked petroleum stocks, or mixtures thereof. The hydrocarbon composition and octane level of the gasoline are not critical. The octane level, (R+M)/2, will generally be above 85. Any conventional gasoline can be used. For example, in the gasoline, hydrocarbons can be replaced by up to substantial amounts of conventional alcohols or ethers conventionally known for use in gasoline. Alternatively, e.g. in countries such as Brazil, the "gasoline" may consist essentially of ethanol.

The gasoline is preferably lead-free, and this may be required by law. Where permitted, lead-free anti-knock compounds and/or valve-seat recession protectant compounds (e.g. known potassium salts, sodium salts or phosphorous compounds) may be present.

Modern gasolines are inherently low-sulphur fuels, e.g. containing less than 200 ppmw sulphur.

In addition to the fatty acid salt of formula I and the co-additive, the gasoline composition may additionally contain one or more corrosion inhibitors, anti-oxidants, dyes, dehazers, metal deactivators, detergents other than a nitrogen-containing detergent (a) (e.g. a polyether amine), carriers other than a polyalphaolefin (b) or a polyoxyalkylene compound (c), diluents and markers.

In gasoline compositions of the present invention, the fatty acid salt is preferably present in an amount in the range 25 to 1000 ppmw, more preferably 25 to 400 ppmw. It is further preferred for the amount to be at

least 50 ppmw. Amounts in the range 50 to 200 ppmw have been found to be very effective.

The co-additive is preferably present in an amount in the range 600 to 1500 ppmw, more preferably 700 to 1500 ppmw, conveniently 700 to 1200 ppmw, based on total composition.

The composition preferably contains at least 400 ppmw, based on total composition, of at least one of (b) and (c). The composition may, for example, contain at least 200 ppmw of (b) and at least 200 ppmw of (c). Amounts of (b) and/or (c) individually in the range 200 to 800, subject to preferred overall maxima for concentration of co-additive are preferred.

For example, a gasoline composition of the invention may contain 200 to 800 ppmw (b) and 200 to 800 ppmw (c), the ratio (b):(c) being in the range 4:1 to 1:4, preferably 3:1 to 1:3, subject to total concentration of (a) (where present), (b) and (c) being in the range 600 to 2000 ppmw, preferably 600 to 1500 ppmw, more preferably 700 to 1500, conveniently 700 to 1200 ppmw. One example, would be 200 ppmw of a polyalphaolefin (b) and 600 ppmw of a polyoxyalkylene compound (c).

In this specification, amounts (concentrations) (ppmw) of fatty acid salt, of co-additive, and of individual components (a), (b) and (c) are of active matter, i.e. exclusive of volatile solvents/diluent materials.

The invention further provides a process for the preparation of a gasoline composition of the invention as defined above which comprises bringing into admixture the gasoline, the fatty acid salt and the co-additive.

If desired, the fatty acid salt, the co-additive, and any additional components such as corrosion inhibitors, anti-oxidants, etc., as listed above, may be co-mixed, preferably together with suitable diluent(s),

in an additive concentrate, and the additive concentrate may be dispersed into gasoline, in suitable quantity to result in a composition of the invention.

The invention also provides a method of operating a spark-ignition engine, which comprises bringing into the combustion chambers of said engine a gasoline composition of the invention as defined above.

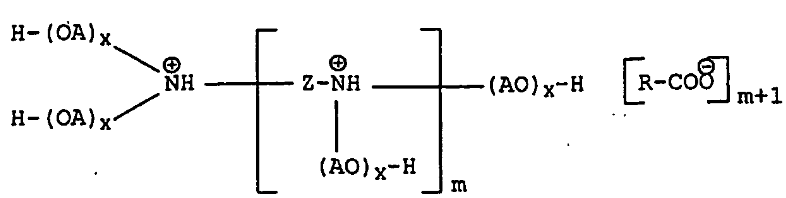
The invention will be further understood from the following illustrative examples, in which, unless otherwise indicated, parts and percentages are by weight, and the temperatures are in degrees Celsius.

In the examples, base fuel used was an unleaded gasoline (95 USG) of RON 95.2, MON 87.2, and having sulphur content (ASTM D 2622-94) 12 ppmw, aromatics content (DIN 51413/T3) of 31.93 v/v, total paraffins content 61.56 v/v, density (DIN 51757/V4) 733.3 kg/m^3, distillation (ISO 3405/88) IBP 38°C, 10% v/v 55°C, 50 % v/v 101°C, 90% v/v 142°C and FBP 169°C.

Fuels were blended with additives by adding additives to base fuel at ambient temperature and homogenising.

The following additives were used:-

"FM" - this was a friction modifier additive in the form of a fatty acid salt of an alkoxylated oligoamine of general formula I



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wherein indices A represent 1,2-propylene groups, variables R represent a mixture of C_{17} moieties such that R-COOH corresponds to a mixture of oleic and linoleic acids in

approximate molar ratio 2:1, Z represents polymethylene groupings of formula $-(CH_2)_n-$, where n is 2 to 6, m is 1, and x is 1, prepared in analogous manner to Example 1 of DE-A-19953651 using a mixture of alpha, omega - C_{2-6} diamines in place of ethylene diamine, and a 2:1 mixture of oleic and linoleic acids in place of oleic acid per se.

"DP" - this was a standard commercial gasoline additive package, containing a polyisobutyleneamine detergent, a synthetic carrier oil and a conventional corrosion inhibitor, corresponding closely to additive package PI of Example 3 of DE-A-19955651. The polyisobutyleneamine detergent was a polyisobutylene monoamine (PIBA) ex BASF, in which the polyisobutylene (PIB) chain has a number average molecular weight of approximately 1000. The synthetic carrier oil was a polyether carrier fluid similar to PCF and PGHE below. The additive package contained about 68% in non-volatile matter, about 27 %w of the package being the PIBA and 40 %w of the package being the polyether carrier fluid.

"PCF" - this is a polyether carrier fluid, similar to PGHE below and to the synthetic carrier oil in "DP" above, being a polyoxypropylene glycol hemiether containing 15 to 30 propylene oxide units prepared using a mixture of alkanols in the C_{8-15} range as initiator (having Mn in the range 1000 to 2000).

"PGHE" - this is a polyether carrier fluid in the form of a polyoxypropylene glycol hemiether (monoether) prepared using a mixture of C_{12-15}

alcohols as initiator having Mn in the range 1200 to 1500 and a kinematic viscosity in the range 72 to 82 mm^2/s at 40°C according to ASTM D 445. "PGHE" contains 17 to 22 propylene oxide units.

"PAO" - this is a polyalphaolefin, being a hydrogenated oligomer of decene-1 having a viscosity at 100°C of 8 x 10^{-6} m^2/s (8 centistokes).

"DET" - this is a 50 %w solution in volatile hydrocarbon solvent of the PIBA component of DP above.

Fuels were tested for acceleration performance using a Ford Zetec 2.0 litre in-line 4 cylinder, 16-valve, multipoint fuel injection, spark-ignition engine, in a bench engine test as follows.

The test is based on a cycle of engine accelerations from 1300 to 4000 rpm as set forth in Table 1:-

Table 1

Cycle Stage	Engine Speed (rpm)	Engine Torque (Nm)	StageTime (s)	Throttle position (%)	Coolant out (tem., °C)	Oil in (temp. °C)
1	1300	20	7	13	100	105
2	1300-4000	140	Variable	100	100	105
3	4000-1300	90	5	13	100	105

Stage time for stage 2 is variable, according to the fuel used, being approximately 8 seconds for base fuel. Acceleration times are measured in Stage 2 for the time interval taken for the engine to pass from 1500 to 3500 rpm.

In the test, a sequence of 40 test cycles is made to establish an average base fuel acceleration time. The fuel is then changed to test fuel (which comprises the same base fuel plus additive(s)), and a sequence of 40 test cycles is made to establish an average test fuel

acceleration time. For each test fuel, there is established an acceleration time benefit as a percentage of the difference between the acceleration times relative to the base fuel time.

Following a test as described above, a purging procedure is carried out, in which the engine is run for 16 hours on base fuel, with three engine oil changes being carried out during that period, in order to ensure that there is no contamination of engine oil by test fuel additives in subsequent tests.

Details of the test fuels and acceleration performance data are given in Table 2 following, wherein Examples 1 to 5 are examples of the invention, and Comp. A to Comp. E are comparative examples:-

Table 2

Example	FM dose (ppmw)	Additive	Additive dose (ppmw)	Additive active matter (ppmw)	Acceleration performance benefit (%)
1	50	DP	1680	1126*	3.090
2	100	DP	1680	1126*	2.753
3	200	DP	1680	1126*	3.840
4	50	PAO	800	800	2.437
5	50	PCF	800	800	2.935
Comp. A	50	DP	525	352**	1.950
Comp. B	800	-	-	-	1.577
Comp. C	-	PIBA	907	454	0.695
Comp. D	-	PGHE	672	672	1.91
Comp. E	-	DP	1680	1126*	2.13

* corresponds to 454 ppmw PIBA plus 672 ppmw polyether carrier fluid
** corresponds to 142 ppmw PIBA plus 210 ppmw polyether carrier fluid

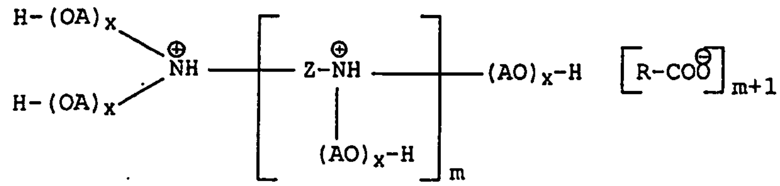
In Example 3 of DE-A-19955651, tests were effected on lubricity of gasolines, including gasolines containing zero additive, 500 mg/kg (ppmw) of additive package P1, and 50 mg/kg (ppmw) of friction modifier product Example 1 of DE-A-19955651 plus 500 mg/kg (ppmw) of additive package P1, using a high frequency reciprocating rig (HFFR) test, in which the gasoline used was a 50% distillation residue of a standard Eurosuper gasoline.

Addition of PI alone (or the similar P2 package) was said to result in frictional wear values of the same order of magnitude as the blank values, whilst addition of 50 ppmw of the friction modifies product of Example 1 gave lower values, and results which were stated to be clearly superior to those resulting from inclusion of 50 ppmw of a prior art lubricity improver.

In the light of the teaching of Example 3 of DE-A-19955651, the acceleration performance findings of the present invention as evidenced by Table 2 above are extremely surprising. In particular, it should be noted that the comparative example Comp. A corresponds very closely to the combination giving the best results in Example 3 of DE-A-19955651.

CLAIMS

1. A gasoline composition comprising a major amount of a gasoline suitable for use in a spark ignition engine; 5 to 1000 ppmw, based on total composition, of a fatty acid salt of an alkoxylated oligoamine of general formula I



(I)

wherein each moiety A independently represents an alkylene group of 2 to 8 carbon atoms, each moiety R independently represents a C_{7-23} alkyl or singly or multiply-unsaturated C_7 to C_{23} alkenyl group, optionally substituted by one or more -OH groups, each moiety Z independently represents a C_{1-8} alkylene group, a C_{3-8} cycloalkylene group, or a C_{6-12} arylene or arylalkylene group, m represents 0 or an integer in the range 1 to 5, and the total of all variables x has a value of 50% to 300% of (m+3); and 600 to 2000 ppmw, based on total composition, of a co-additive selected from the group consisting of (a) a nitrogen-containing detergent containing a hydrocarbyl group having a number average molecular weight (Mn) in the range 750 to 6000, (b) a polyalphaolefin having a viscosity at 100°C in the range $2x10^{-6}$ to $2x10^{-5}$ m^2/s (2 to 20 centistokes), being an oligomer containing 18 to 80 carbon atoms derived from at least one alphaolefinic monomer containing from 8 to 16 carbon atoms; (c) a polyoxyalkylene compound selected from glycols, mono- and diethers thereof, having number

average molecular weight (Mn) in the range 400 to 3000; and mixtures of any two or all three of (a), (b) and (c).

2. Composition according to Claim 1 wherein in formula I each moiety A independently represents a 1,2-ethylene, 1,2-propylene or 1,2-butylene group, each moiety R independently represents a singly or multiply-unsaturated C_{15-19} alkenyl group, optionally substituted by one or more -OH groups, each moiety Z is a polymethylene group of formula $-(CH_2)_n-$, where n is 2 to 6, m is 1 or 2, and the total of all variables x has a value of 75% to 125% of (m+3).

3. Composition according to Claim 1 or 2 wherein the nitrogen-containing detergent (a) is a hydrocarbyl amine or formula R^1-NH_2, wherein R^1 represents a group R_2 or a group R^2-CH_2- and R^2 is a hydrocarbyl group having a number average molecular weight in the range 950 to 1350.

4. Composition according to Claim 1, 2 or 3 wherein the poyalphaolefin (b) is derived from an alphaolefinic monomer containing from 8 to 12 carbon atoms, and has viscosity at 100°C in the range $6x10^{-6}$ to $1x10^{-5}$ m^2/s (6 to 10 centistokes).

5. Composition according to any one of Claims 1 to 4 wherein the polyoxyalkylene compound (c) has the formula II

$$R^3—O—\left(R^5\right)_p—O—R^4 \qquad \text{(II)}$$

wherein R^3 and R^4 independently represent hydrogen atoms or C_{1-40} hydrocarbyl groups, each R^5 independently represents a C_{2-8} alkylene group and n is such that M_2 of the polyoxyalkylene compound is in the range 700 to 2000.

6. Composition according to any one of Claims 1 to 5, wherein the fatty acid salt is present in an amount in the range 25 to 400 ppmw, based on total composition.

7. Composition according to any one of Claims 1 to 6, wherein the co-additive is present in an amount in the range 700 to 1500 ppmw, based on total composition.

8. Composition according to any of Claims 1 to 7 which contains at least 400 ppmw, based on total composition, of (b) and/or (c).

9. A process for the preparation of a gasoline composition according to any one of Claims 1 to 8 which comprises bringing into admixture the gasoline, the fatty acid salt and the co-additive.

10. A method of operating a spark-ignition internal combustion engine, which comprises bringing into the combustion chambers of said engine a gasoline composition according to any one of Claims 1 to 8.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP 03/01397

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C10L1/14

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C10L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ, API Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 199 55 651 A (BASF AG) 23 May 2001 (2001-05-23) cited in the application the whole document ---	1-10
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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
26 May 2003	05/06/2003

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Keipert, O

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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A	EP 0 588 429 A (SHELL INT RESEARCH) 23 March 1994 (1994-03-23) the whole document -----	1-10

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